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Self-striking red glass fabrication at low temperature using gold nanoparticles

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Abstract

This research is intended to determine the appropriate condition for gold ruby glass production at low temperature. The effects of reducing agent (SnO_2 and SeO_2) concentrations on coloration of gold nanoparticle (AuNPs) in glass samples have been investigated. The glasses with chemical compositions of SiO_2, B_2O_3, Al_2O_3, Na_2O, CaO, K_2O, Sb_2O_3, SnO_2, SeO_2 and AuNPs were fabricated by conventional melt-quench technique at 1,200°C in normal atmosphere. The results found that the red glasses were obtained by SnO_2 with concentration of 0.5 wt% and SeO_2 with concentration of 0.05 wt%. The color of glasses were confirmed by UV-visible spectrophotometer in the wavelength range 300-1100 nm and color coordinate in CIE L*a*b* system. Moreover, the color of glasses were obtained immediately when took the glass out of furnace without second heat treatment.

1. Introduction

It is well known that the red glass from gold nanoparticles (AuNPs) is interesting in glass industrial production because it is the most precious and popular among all red pigments [1-3]. The glass doped with gold nanoparticles is known as “gold ruby glass” or “cranberry glass”. The red coloration is caused by the optical phenomenon exhibited by gold nanoparticles is surface plasmon resonance (SPR) [4,5]. SPR is the collective oscillation of the electrons of conduction electrons that are excited by the interaction of electromagnetic wave. The SPR absorption band of this interaction is strongly dependent on the size and shape distribution of nanoparticles which indicate to the optical absorption spectra in the UV-vis region and display the desired color [6]. Its found that spherical AuNPs with size distribution is between few and about 50 nm has the maximum absorption peak locate at 520-530 nm and there are red shift with the increasing diameters of gold nanoparticles [7]. Moreover, the factors affecting controlling the size distribution is very important for the coloration in glass such as temperature and soaking time in melting process, duration and temperature of the annealing step, reducing agent types and the composition of the base glass. These factors particularly affect on the presence of characteristic elements influencing the precipitation of the metal nanoparticles [8].

In glass production, the red glass is not easy to produce because of coloration control difficulties, it need a second heat treatment known as “striking”. Nevertheless, the gold ruby glass can be developed to the suddenly striking red color without a second heat treatment, this process is called “self-striking” [9]. Recently, it has been reported the red glass preparation at high temperature for self-striking process [10,11]. However, the fuel cost for glass preparation at high melting temperature is high, so glass production at low melting temperature is alternative way for saving cost. In the present work, tin oxide (SnO_2) and selenium oxide (SeO_2) were selected as a reducing agent for self-striking process at low temperature. In glass matrix, the diffused Au^+ can be reduced to neutral atomic state (Au^0) by reducing agents and then the gold atoms agglomerate to form nanoclusters of appropriate

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size and produce the desired color [12]. Therefore, the effect of SnO_2 and SeO_2 on glass coloration from AuNP in self- striking process at low melting temperature were investigated.

2. Experimental

The glass samples containing a reducing agents with different concentration were prepared in composition of SiO_2, B_2O_3, Al_2O_3, Na_2O, CaO, K_2O, Sb_2O_3, SnO_2, SeO_2 and AuNPs as shown in Table 1 and 2. All the chemical compositions were finely powder while AuNPs was solution (diameter = 16 nm). The whole of composite were mixed in a high purity alumina crusible (each batch weighs for 20 g). Then the batches were melted by placing them in an electrical furnace at 1200°C for 3 h. After complete melting, these melts were quenched in air by pouring between preheated stainless steel plates. The quenched glasses were annealed at 500°C for 3 h to reduce thermal stress, and cooled down to room temperature. Finally, all glass samples were cut and polished for further investigation.

The densities (ρ) were measured by the Archimede's method using xylene as immersion fluid. The corresponding molar volume (V_M) was calculated using the relation, $V_M = M_T/\rho$, where M_T is the total molecular weight of the multi-component glass. Refractive index (RI) was measured at room temperature using a DR-M2 refractometer with a refractometer fluid $n_D \leq 1.65$ and the sodium vapor lamp as the light source (539 nm). The UV-Vis-NIR Spectrophotometer (UV-3600) used to measure optical spectra of glass sample and CIE L*a*b* color coordinate calculation. the excitation and emission spectra of the samples measured by luminescence spectrometer (Cary Spectrophotometer).

3. Results and discussion

3.1 Glass samples

The glass samples with different reducing agents are illustrated in Figures 1 and 2. The undoped glass shows the purple color that was obtained from AuNPs with diameter larger than 50 nm and the color of the glass changed with the increase of reducing agent content. The glasses with SnO_2 concentrations of 0.1 to 0.4 wt% and SeO_2 concentrations of 0.01 to 0.04 wt% show purple color while red color was occurred from SnO_2 concentrations at 0.5 and SeO_2 concentrations at 0.05 wt%. It is important to emphasize that the red color glasses can be obtained immediately when the glass is taken out of the electrical furnace. The color significantly changes from the purple to red color with increasing SnO_2 and SeO_2 concentration. The results reflect that SnO_2 and SeO_2 have an effect on the coloration of the glass in which may relates with the nucleation of gold nanoparticles [11].

Table 1. Chemical compositions of glass samples with different SnO_2 concentration.

Concentration of SnO_2	Glass composition (wt%)								
	SiO_2	Na_2O	CaO	K_2O	Sb_2O_3	Al_2O_3	B_2O_3	AuNPs	SnO_2
0.0	48.92	25	6	10	0.05	2.5	7.5	0.03	-
0.1	48.82	25	6	10	0.05	2.5	7.5	0.03	0.1
0.2	48.72	25	6	10	0.05	2.5	7.5	0.03	0.2
0.3	48.62	25	6	10	0.05	2.5	7.5	0.03	0.3
0.4	48.52	25	6	10	0.05	2.5	7.5	0.03	0.4
0.5	48.42	25	6	10	0.05	2.5	7.5	0.03	0.5

Table 2. Chemical compositions of glass samples with different SeO_2 concentration.

Concentration of SeO_2	Glass composition (wt%)								
	SiO_2	Na_2O	CaO	K_2O	Sb_2O_3	Al_2O_3	B_2O_3	AuNPs	SeO_2
0.00	48.92	25	6	10	0.05	2.5	7.5	0.03	-
0.01	48.91	25	6	10	0.05	2.5	7.5	0.03	0.01
0.02	48.90	25	6	10	0.05	2.5	7.5	0.03	0.02
0.03	48.89	25	6	10	0.05	2.5	7.5	0.03	0.03
0.04	48.88	25	6	10	0.05	2.5	7.5	0.03	0.04
0.05	48.87	25	6	10	0.05	2.5	7.5	0.03	0.05

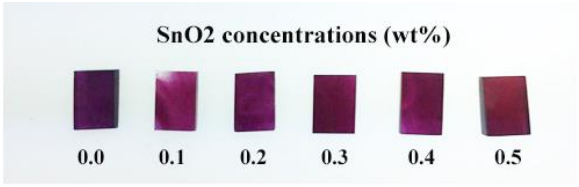

Figure 1. The glass samples with different SnO_2 concentration.

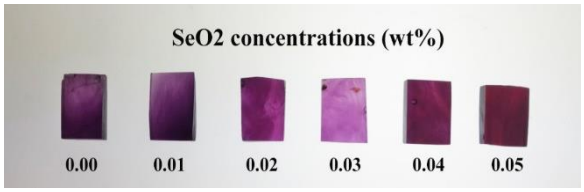

Figure 2. The glass samples with different SeO_2 concentration.

3.2 Density, molar volume and refractive index

The obtained value of density and molar volume are shown in Figures 3 and 4. The density of glass samples with SnO_2 concentrations from 0.0 to 0.5 wt% are within the range of 2.5682-2.5820 g/cm^3 and the molar volume values are in the range of 31.3898- 31.4259 cm^3/mol. While the density of glass samples with SeO_2 concentrations from 0.00 to 0.05 wt% are within the range of 2.5682-2.5820 g/cm^3 and the molar volume values are in the range of 31.3898- 31.4259 cm^3/mol. It has been observed that there is no effect of SnO_2 and SeO_2 concentration on these parameters. Actually, SnO_2 and SeO_2 are heavier than SiO_2 and the glass density should be increased with increasing of SnO_2 and SeO_2 content and then affect to molar volume. From this result, the dependence of density and molar volume with SnO_2 and SeO_2 concentration may be come from losing ratio and also due to the volatilization of low melting point component such as Na_2O and K_2O when glass were melted at high temperature [13]. Moreover, the refractive index value of glass samples with SnO_2 and SeO_2 concentration are in the range of 1.6561-1.6568 and 1.6558-1.6565, respectively as shown in Fig 5 and 6. It has been found that there is no effect of SnO_2 and SeO_2 concentration on such parameters. Theoretically, the refractive index is a function of density and these results maintain this trend. According to classical dielectric theory, the refractive index depends on density and on polarizability of the atom in a given material [14]. The density and the refractive index should be increased when high density component added into the glass structure. However, in this work, it is found that they are independent of SnO_2 and SeO_2 concentration because of the volatilization of low melting point component.

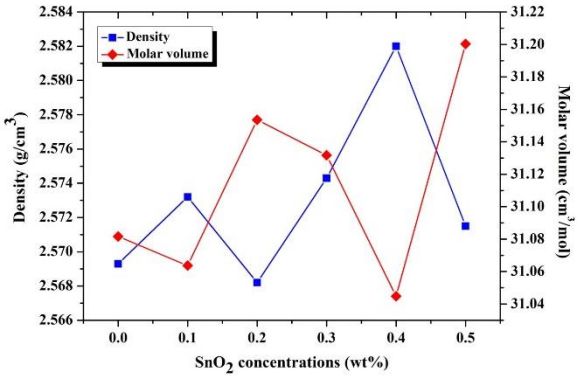

Figure 3. The density and molar volume of glass samples with different SnO_2 concentration.

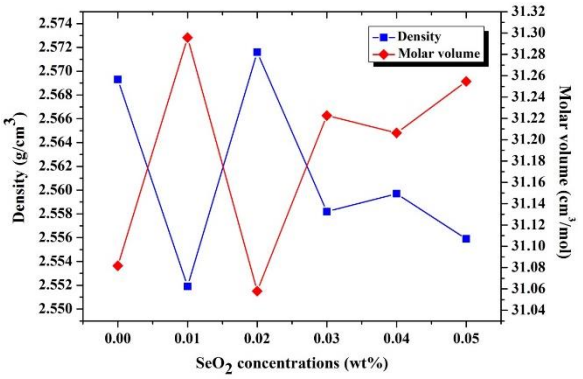

Figure 4. The density and molar volume of glass samples with different SeO_2 concentration.

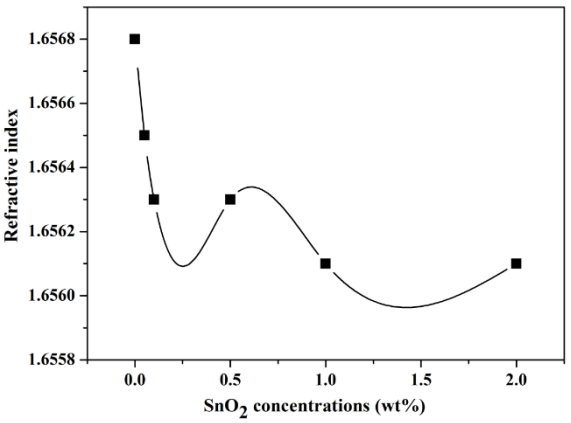

Figure 5. The refractive index of glass samples with different SnO_2 concentration.

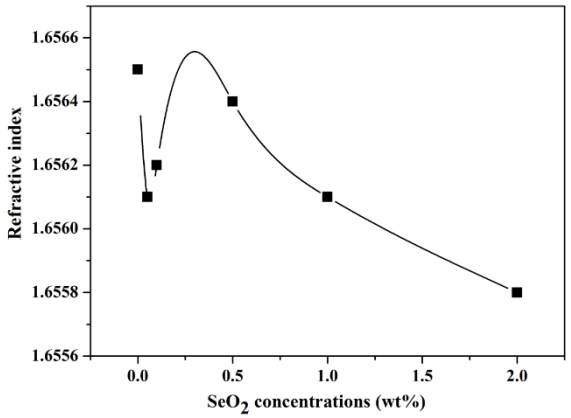

Figure 6. The refractive index of glass samples with different SeO_2 concentration.

3.3 Absorption spectra and CIE L*a*b* system

The absorption spectra of glass samples are illustrated in Figures 7 and 8. The optical absorption spectra were recorded in the range 200-1100 nm at room temperature. It has been found that the absorption peak of undoped glass has predominant peak around 555-565 nm produces purple color. The glass samples with SnO_2 concentrations from 0.0 to 0.4 wt% are observed in the absorption spectra with peaks around 540-560 nm and shown purple color. The absorption broad band around 530 nm is obtained from 0.5 wt% concentrations of SnO_2 and display red color. The absorption spectra of glass samples doped with SeO_2 similar to SnO_2 result. Within the concentration of SeO_2 from 0.00 to 0.04 wt%, a predominant broad band locate around 550-560 nm and shown purple color. The glass sample with SeO_2 concentration at 0.05 wt% shows the absorption board bands around 530 nm and produce red color. The result can be explained by the acceleration of the kinetics of the formation of AuNPs by using SnO_2 and SeO_2 as catalyst [8].

$$Sn^{4+} + 2O^{2-} \rightarrow Sn^{2+} + O_2 \quad (1)$$

$$Sn^{2+} + 2Au^{+} \rightarrow 2Au^{0} + Sn^{4+} \quad (2)$$

It implies that the Au^{+} ions can be reduced to Au^{0} atom by the addition of SnO_2 and SeO_2. After that, the gold atoms agglomerate to condensation nuclei for the nanoparticles with appropriate size in glass matrix. However, the absorption peak position depends on the size and shape distributions of AuNPs. We compared the absorption spectra with literatures, and found that the size distribution of spherical AuNP which is in range of a few and about 50 nm, exhibits the absorption peak at 520-530 nm and display a red color in glass [15,16]. This result is corresponding with published literatures.

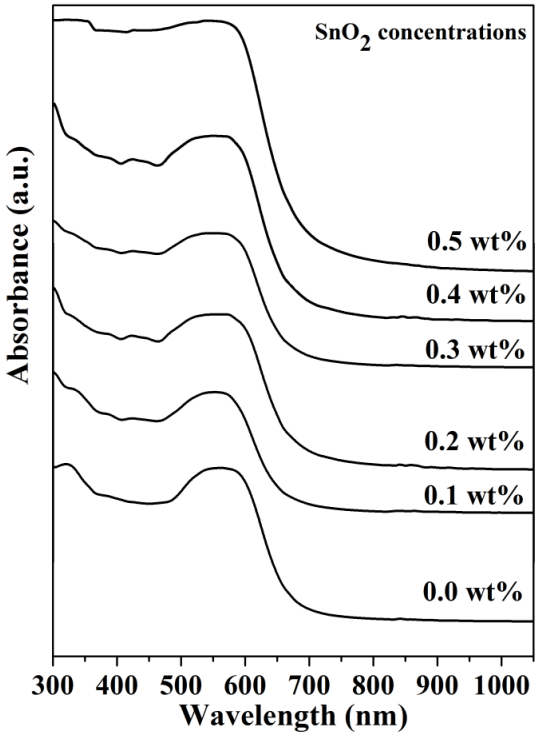

Figure 7. The absorption spectra of glass samples with different SnO_2 concentration.

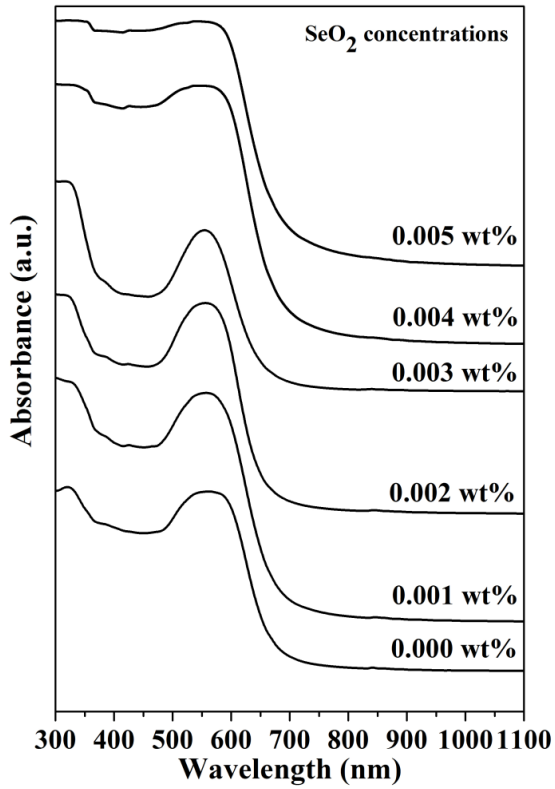

Figure 8. The absorption spectra of glass samples with different SeO_2 concentration.

Moreover, the absorption spectra were consistent the color coordinate in CIEL*a*b* system, as shown in Figures 9 and 10. Therefore, glass samples with SnO_2 at 0.5 wt% and SnO_2 at 0.5 wt% are the optimize concentration for a preparation of red glass at low temperature by self-striking process.

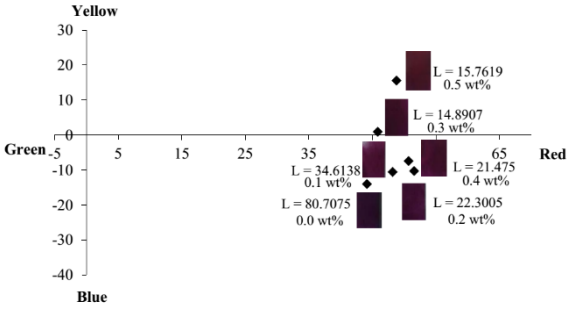

Figure 9. The CIE L*a*b* color scale of glass samples with different SnO_2 concentration.

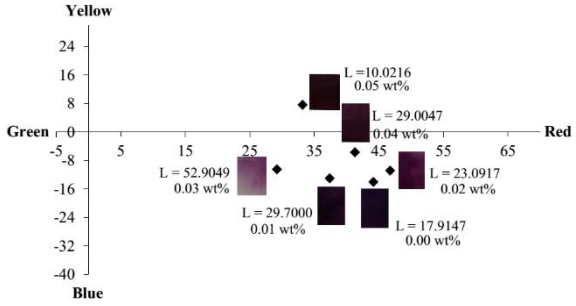

Figure 10. The CIE L*a*b* color scale of glass samples with different SeO_2 concentration.

4. Conclusions

In this work, SnO_2 and SeO_2 were selected as a reducing agent in self-striking process at low temperature for red glass production. The concentrations of SnO_2 in present glasses were varied from 0.0 to 0.5 wt%, while SeO_2 in range 0.00 to 0.05 wt%. The results show that the density and the molar volume are not affected with increasing of SnO_2 and SeO_2 concentrations. The absorption peak locates around 530 nm and show red color with SnO_2 at 0.5 wt% and SeO_2 at 0.05 wt%. The color coordinate in CIEL*a*b* system confirmed color of glasses.

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